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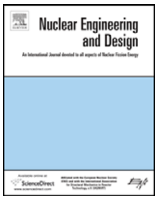

Uranium–zirconium hydride fuel properties

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ABSTRACT

Properties of the two-phase hydride $U_{0.3}ZrH_{1.6}$ pertinent to performance as a nuclear fuel for LWRs are reviewed. Much of the available data come from the Space Nuclear Auxiliary Power (SNAP) program of 4 decades ago and from the more restricted data base prepared for the TRIGA research reactors some 3 decades back. Transport, mechanical, thermal and chemical properties are summarized. A principal difference between oxide and hydride fuels is the high thermal conductivity of the latter. This feature greatly decreases the temperature drop over the fuel during operation, thereby reducing the release of fission gases to the fraction due only to recoil. However, very unusual early swelling due to void formation around the uranium particles has been observed in hydride fuels. Avoidance of this source of swelling limits the maximum fuel temperature to ∼650 °C (the design limit recommended by the fuel developer is 750 °C). To satisfy this temperature limitation, the fuel-cladding gap needs to be bonded with a liquid metal instead of helium. Because the former has a thermal conductivity ∼100 times larger than the latter, there is no restriction on gap thickness as there is in helium-bonded fuel rods. This opens the possibility of initial gap sizes large enough to significantly delay the onset of pellet-cladding mechanical interaction (PCMI). The large fission-product swelling rate of hydride fuel (3× that of oxide fuel) requires an initial radial fuel-cladding gap of ∼300 m if PCMI is to be avoided. The liquid-metal bond permits operation of the fuel at current LWR linear-heat-generation rates without exceeding any design constraint. The behavior of hydrogen in the fuel is the source of phenomena during operation that are absent in oxide fuels. Because of the large heat of transport (thermal diffusivity) of H in ZrH_x, redistribution of hydrogen in the temperature gradient in the fuel pellet changes the initial H/Zr ratio of 1.6 to ∼1.45 at the center and ∼1.70 at the periphery. Because the density of the hydride decreases with increasing H/Zr ratio, the result of H redistribution is to subject the interior of the pellet to a tensile stress while the outside of the pellet is placed in compression. The resulting stress at the pellet periphery is sufficient to overcome the tensile stress due to thermal expansion in the temperature gradient and to prevent radial cracking that is a characteristic of oxide fuel. Several mechanisms for reduction of the H/Zr ratio during irradiation are identified. The first is transfer of impurity oxygen in the fuel from Zr to rare-earth oxide fission products. The second is the formation of metal hydrides by these same fission products. The third is by loss to the plenum as H_2.

The review of the fabrication method for the hydride fuel suggests that its production, even on a large scale, may be significantly higher than the cost of oxide fuel fabrication.

1. Introduction

The history of uranium–zirconium hydride as a fuel or zirconium hydride as a moderator for nuclear reactors goes nearly as far back as that of oxide fuels. This fuel/moderator occupies a niche in reactor technology with a number of proposed designs and fewer actual units. Among the former are the hydride-moderated boiling-water superheat reactor investigated by the National Aeronautics Administration as early as 1960 (Gylfe et al., 1960).

Despite the paucity and age of available data on U,Zr hydrides, such information is essential for determining whether any fuel-related constraints are likely to limit performance as a power-reactor fuel. Potential limiting factors include maximum temperature, internal rod pressure rise due to fission-gas release, cladding strain from pellet-cladding mechanical interaction (PCMI) and waterside corrosion.

The purpose of this contribution is to review the history of hydride-fueled reactors in order to extract information required for the design of a power-reactor fuel using this material and to

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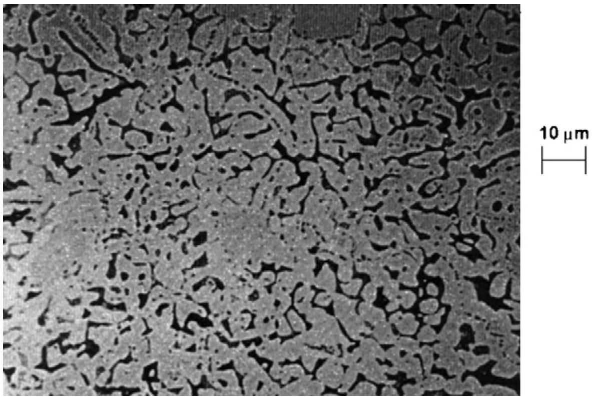

Fig. 1. Photomicrograph of $(U_{0.31}Zr)H_{1.6}$. The black areas are (probably) uranium metal; the gray region is $ZrH_{1.6}$. The white dots are unidentified (from West et al., 1986).

Table 1
Comparison of reactor fuel features.

Reactor type	SNAP	LWR-hydride	LWR-oxide
Weight fraction uranium in fuel, w_U	0.1	0.45	0.88
Enrichment in ^{235}U, e	0.93	0.125	<0.05
U/Zr atom ratio in fuel, y	0.043	0.31	–
H/Zr atom ratio, x	1.68–1.83	1.6	–
Fuel density, ρ, g/cm^3	6.1	8.3	10.5
Uranium density, ρ_U, g U/cm^3	0.6	3.7	9.3
^{235}U density = $e\rho_U$	0.56	0.46	0.46
Fuel pellet (diameter × length, in cm)	1.4 × 35.6	1.2 × 1.5	1.2 × 1.5
Gap filler	He, 0.1 atm	Liquid metal	He, 20 atm
Cladding material	Hastelloy	Zircaloy (?)	Zircaloy-2
Peak fuel temperature, °C	850	550	1450
Peak linear heat rate, kW/m	~14	37.5	37.5
Peak cladding temperature, °C	~750	~350	~350
Average burnup	1.4×10^{-3} FIMA[a]		60 MWd/kgU

[a] Fissions per initial metal atoms; also called "metal atom fraction fissioned".

summarize what is known about the properties of uranium–zirconium hydride pertinent to its use as a fuel for LWRs.

This section reviews the work done on the early Space Nuclear Auxiliary Power (SNAP) project starting in the 1960s, about the same time that the popular TRIGA research reactor was marketed to universities and nuclear laboratories. Section 2 presents most of what is known about the transport, thermal, mechanical and chemical properties of ZrH_x. Section 3 gives information extracted from 30 to 40-year-old reports on the irradiation properties of hydride fuel. A summary of these properties up to 1981 is given by Simnad (1981). Section 4 discusses the in-reactor chemical/materials performance of this material. Section 5 presents a comparison of oxide and hydride fuels and Section 6 reviews batch production methods of $(U,Zr)H_x$. Section 7 calculates the closure of the fuel-cladding gap as a function of burnup.

1.1. Measures of composition

As shown in Fig. 1, the hydride fuel is a two-phase mixture consisting of a continuous $ZrH_{1.6}$ matrix in which small particles of uranium metal are embedded.

Hydride fuel contains three elements, and a variety of units have been used to denote its composition. One convenient designation is $(U_yZr)H_x$, where x is the H/Zr atom ratio and y is the U/Zr atom ratio:

$$y = \frac{N_U}{N_{Zr}} \tag{1}$$

where

$$N_{Zr} = \frac{6.02 \times 10^{23} \rho (1 - w_U)}{91.2} \tag{2}$$

ρ is the density of the fuel and w_U is the weight fraction of uranium. Weight fraction and atom ratio are related by

$$y = \frac{91.2}{M_U} \times \frac{w_U}{1 - w_U} \tag{3}$$

where M_U is the atomic weight of uranium:

$$M_U = 235e + 238(1 - e) \tag{4}$$

and e is the enrichment.The room-temperature density of the uranium phase is $\rho_U^o = 19.9$ g/cm^3, and that of the zirconium hydride phase is (Simnad, 1981):

$$\rho_{ZrH_x} = (0.154 + 0.0145x)^{-1} \ \text{g/cm}^3 \quad x < 1.6 \tag{5a}$$

$$\rho_{ZrH_x} = (0.171 + 0.0042x)^{-1} \ \text{g/cm}^3 \quad x > 1.6 \tag{5b}$$

The density of the two-phase mixture that constitutes the hydride is

$$\rho = \left(\frac{w_U}{\rho_U^o} + \frac{1 - w_U}{\rho_{ZrH_x}} \right)^{-1} \tag{6}$$

The uranium density of the fuel, given by

$$\rho_U = w_U \rho \tag{7}$$

is a key property as it dictates the enrichment of the uranium required to achieve a desirable cycle length. The upper limit is $w_U = 0.45$ (Chesworth and West, 1985).The molecular weight of the hydride is defined as the mass in grams per mole of zirconium, or:

$$M = yM_U + 91.2 + x \tag{8}$$

1.2. The SNAP reactors

A hydride-fueled reactor that received more than passing attention was developed for the SNAP program at Atomics International under the auspices of the Atomic Energy Commission, the predecessor of the current Dept. of Energy (Lillie et al., 1973). Six reactors with thermal outputs ranging from 50 kW to 1 MW were built and operated, and one was placed in earth orbit. A substantial body of experimental irradiation tests was reported in this program, many of which have a bearing on the hydride-fueled LWR that is the subject of this issue. Table 1 compares the pertinent characteristics of three reactor types: the SNAP reactor, the LWR-hydride reactor (Shuffler et al., this issue-a) and the LWR-oxide reactor (a standard BWR).

Noteworthy in Table 1 is the wide range of uranium contents of the fuels. Hydride fuels operate most reliably with low U concentrations, which is the reason for the choice of 10 wt% U, corresponding to a uranium density of 0.6 g U/cm^3, in the SNAP reactors. To produce acceptable amounts of nuclear energy with so little uranium per unit volume, the enrichment is 93% ^{235}U. In order to attain the same ^{235}U density of the fresh fuel as in an oxide-fueled core, the enrichment of the LWR-hydride fuel would have to be scaled according to the total uranium density, or:

$$e(\text{LWR} - \text{hydride}) = e(\text{LWR} - \text{oxide}) \times (9.3/3.7).$$

To match a 5%-enriched oxide fuel in this regard, the hydride fuel for LWR use would require 12.5% enriched uranium. This enrichment is higher than the current regulatory limit of 5%, but is below the LEU limit of 20%.

The power density of SNAP fuel was only ~1/4 that of current LWR oxide fuel. But then, the SNAP reactors were not designed